Numerical study of transport pathways of ^{137}Cs from forests to freshwater fish living in mountain streams in Fukushima, Japan

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Highlights

- Cs-137 concentrations in some freshwater fish in Fukushima remain high, so a model was developed to assess uptake pathways.
- Model assumed three export pathways from forests which supply soluble ^{137}Cs to rivers were relevant for uptake by fish.
- Pathways were direct litter fall into rivers, lateral inflow from litter, and transfer from soil via runoff and groundwater.
- Data on ^{137}Cs in forests, river water and freshwater fish measured across Fukushima were used for model calibration.
- Fish ^{137}Cs concentrations predicted to reach steady state after around 10 y due to equilibration of ^{137}Cs cycle in forests.

Abstract

The accident at the Fukushima Dai-ichi Nuclear Power Plant in 2011 released a large quantity of radiocesium into the surrounding environment. Radiocesium concentrations in some freshwater fish caught in rivers in Fukushima Prefecture in October 2018 were still higher than the Japanese limit of 100 Bq kg^{-1} for general foodstuffs. To assess the uptake of ^{137}Cs by freshwater fish living in mountain streams in Fukushima Prefecture, we developed a compartment model for the migration of ^{137}Cs on the catchment scale from forests to river water. We modelled a generic forest catchment with Fukushima-like parameters to ascertain the importance of three export pathways of ^{137}Cs from forests to river water for the uptake of ^{137}Cs by freshwater fish. The pathways were direct litter fall into rivers, lateral inflow from the forest litter layer, and lateral transfer from the underlying forest soil. Simulation cases modelling only a single export pathway did not reproduce the actual trend of ^{137}Cs concentrations in river water and freshwater fish in Fukushima Prefecture. Simulations allowing a combined effect of the three pathways reproduced the trends well. In the latter simulations, the decreasing trend of ^{137}Cs in river water and freshwater fish was due to a combination of the decreasing trend in the forest leaves/needles and litter compartments, and the increasing trend in soil. The modelled ^{137}Cs concentrations within the forest compartments were predicted to reach an equilibrium state at around ten years after the fallout due to the equilibration of ^{137}Cs cycling in forests. The model suggests that long term ^{137}Cs concentrations in freshwater fish in mountain streams will be controlled by the transfer of ^{137}Cs to river water from forest organic soils.

1. Introduction

The accident at the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) in 2011 released a large quantity of radionuclides into the environment (Saito and Onda, 2015). Cesium-134 and ^{137}Cs, with half-lives of 2.1 and 30 years respectively, remain as the main radionuclides within the environment (Evrard et al., 2015). In the first few years following the accident, radiocesium concentrations in most agricultural and marine fish products dropped quickly (Wada et al., 2016b; Tagami and Uchida, 2016). However, as of October 2018, radiocesium concentrations in some freshwater fish caught within Fukushima Prefecture remain higher than the Japanese limit of 100 Bq kg^{-1} for general foodstuffs (Wada et al., 2016a, 2019; Ministry of Agriculture, Forestry and Fisheries, 2018a; Fukushima Prefecture, 2018ab). Fish caught both inside and outside the evacuation zone surrounding the FDNPP have exceeded this limit (Table 1). Examples include ayu (*Plecoglossus altivelis*), caught with up to about 2 kBq kg^{-1} ^{137}Cs in rivers near the FDNPP in 2016, masu salmon (*Oncorhynchus masou*) (resident form), e.g. 126 Bq kg^{-1} ^{137}Cs in a sample caught in a tributary of the Abukuma river in April 2018, and white-spotted char (*Salvelinus leucomaenis*), 195 Bq kg^{-1} ^{137}Cs in a sample caught in a tributary of the Abukuma river in October, 2018.

Freshwater fish caught in lakes and rivers traditionally make up a significant part of the Japanese diet. It is important therefore to understand the mechanisms of radiocesium export from overland to rivers which influence radiocesium concentrations in freshwater fish. It is thought that radiocesium exported from forests into the aquatic environment is the main source of radiocesium taken up by some species of freshwater fish living in mountain streams in Fukushima Prefecture (e.g. Murakami et al., 2014), as forests cover a major part of the contaminated catchments (c.a. 64% by area, Yamaguchi et al., 2014).

The main mechanism of export of radiocesium from forests to rivers is overland erosion and discharge of radiocesium-bearing soil particles into watercourses, essentially during

heavy rainfall events (Ueda et al., 2013; Nagao et al., 2013; Yamashiki et al., 2014). Around 0.05 – 0.19% of the radiocesium inventory of Fukushima river catchments discharges into rivers annually due to soil erosion (Niizato et al., 2016). This mechanism is not however considered to be the main source of the radiocesium taken up by freshwater fish, as this radiocesium is strongly absorbed to soil particles and barely desorbs on the timescale of rainfall events (Murota et al., 2016; Mukai et al., 2018).

Dissolved radiocesium in river water is the most relevant fraction for biological availability (International Atomic Energy Agency, 2010). The dissolved fraction comprises around 12 – 91% of all radiocesium discharged through rivers under base flow conditions (Ochiai et al., 2015; Eyrolle-Boyer et al., 2016; Tsuji et al., 2016). It has been suggested that degradation of organic matter in forest litter could provide a source of dissolvable radiocesium for input into rivers (Sakuma et al., 2018). Submerged litter in rivers is considered to be another source of dissolvable ^{137}Cs in aquatic ecosystems (Sakai et al., 2015; 2016ab). In the field of nutrient cycling in freshwater ecosystems, previous studies have proved that direct litter fall and litter carried by surface runoff contributes to organic matter input into rivers (e.g. Kochi et al., 2010; Tonin et al., 2017). Thus the literature on inflows of radiocesium and other materials into rivers implies that there should be several radiocesium transport pathways from forests to rivers.

The objective of this study was to examine the ^{137}Cs transport pathways from forests that affect ^{137}Cs concentrations in freshwater fish living in mountain streams in Fukushima Prefecture. A compartment model was developed that simulates dissolved ^{137}Cs circulation on the catchment scale between forests, rivers and freshwater fish. It was assumed for the model that only three transport pathways from forests leading to the input of dissolved ^{137}Cs into river water were relevant for uptake by freshwater fish. The pathways were direct litter fall into rivers, lateral inflow from litter layers in forests, and lateral inflow from underlying forest

soils. The pathways encompass the input of litter, and the input of dissolved ^{137}Cs produced by leaching from organic matter and desorption from soil minerals via surface runoff and groundwater flows.

We applied the model to a generic Fukushima-type forest catchment to evaluate the relative importance of the three transport pathways from forests for ^{137}Cs uptake by freshwater fish. Datasets on ^{137}Cs cycling in several forests in Fukushima Prefecture were used for model calibration and validation. Measurements for ^{137}Cs concentrations in river water and freshwater fish from the Abukuma River and other coastal catchments in Fukushima Prefecture were used for discussion purposes and to provide comparisons for the simulation results. Note only ^{137}Cs was modelled as the transfer dynamics of ^{134}Cs were assumed to be identical.

2. Methods

2.1 Compartment model

A new compartment model was developed for this study to assess, on the river catchment scale, ^{137}Cs transfer between leaves/needles, branches, bark, sap wood, heart wood, litter layer and soil in forests, discharge into rivers, and uptake by freshwater fish. For each compartment *i*, the generic mass balance equation for ^{137}Cs inventory A_i (Bq) is

$$\frac{dA_i}{dt} = -\left\{\lambda_p + \sum_{j=1, i\neq j}^{n} \lambda_{ij}\right\} A_i + \sum_{j=1, i\neq j}^{n} \lambda_{ji} A_j \qquad (1)$$

where t is time (yr), λ_p is the physical decay constant of ^{137}Cs (yr^{-1}) and λ_{ij} is the transfer rate from compartment i to compartment j (yr^{-1}).

Figure 1 shows a schematic of the compartments in the model. Each forest type

comprised seven compartments: leaves/needles, branches, bark, sap wood, heart wood, litter layer and soil. The cycling of radiocesium in forests in Fukushima Prefecture broadly depends on whether the stand is deciduous or coniferous (Imamura et al., 2017). Deciduous and coniferous forests were therefore represented as separate entities in the model, i.e. each type was given its own group of seven forest compartments (Fig. 2).

A river compartment was linked to the leaves/needles, litter layer and soil compartments in both deciduous and coniferous forests. These links model implicitly the following physical processes. The link from the leaves/needles compartment models litter fall directly into rivers and its subsequent breakdown which leaches ^{137}Cs into river water. The link from the litter layer models litter transferred by surface runoff into rivers and its subsequent breakdown, and also leaching from litter on the forest floor which inputs dissolved ^{137}Cs into surface runoff and groundwater flows, and on into rivers. The link from the soil compartment models ^{137}Cs leaching from underlying forest soils into surface water and groundwater and subsequent flow into rivers. Note submerged litter was not modelled as a separate compartment, as ^{137}Cs leaching from submerged litter was considered to occur on a timescale faster than that relevant for the compartment model. This was based on experiments showing ^{137}Cs leaching from submerged litter occurs over the timescale of a few days (Sakai et al., 2015).

A downstream compartment was connected to the river compartment and used as a sink for the model catchment. Thus in total there were sixteen main compartments in the model. An interaction matrix for the compartments is shown in Fig. 2. The compartments and processes in our model are similar to existing forest models (e.g., International Atomic Energy Agency, 2002; Nishina and Hayashi, 2015; Nishina et al., 2018; Thiry et al., 2018). The compartments were chosen to coincide with the main types of monitoring data available for Fukushima Prefecture from previous studies (Ministry of Agriculture, Forestry and Fisheries, 2018b; Komatsu et al., 2016; Imamura et al., 2017).

A fish compartment was connected to the river compartment to model ^{137}Cs uptake by freshwater fish. As previous studies reported, freshwater fish take up ^{137}Cs not from water but mainly from food, and the apparent transfer factor, i.e. the ratio of ^{137}Cs concentration in fish to that in water, depends on the trophic level (e.g., Rowan et al., 1998; Tuovinen et al., 2012; Sundbom et al., 2003). On the other hand, commonly used bioaccumulation models assume that radionuclide concentrations in aquatic organisms are in equilibrium with a reference medium in the surrounding environment such as water or sediment (International Atomic Energy Agency, 2010). This assumption means models can be significantly simplified by not having to model full details of the food chain. Moreover detailed food chain models are often unjustified due to large uncertainties in the transfer parameters between food chain elements. Figure 3 shows a strong relationship between ^{137}Cs concentrations in water and in fish in Fukushima, which justifies the idea that river water can be used as a reference medium.

In this study, the transfer model from river water to fish was identical for all species. The model represented all the dynamic processes of food chains (uptake to fish via plankton, worms etc.) implicitly by using the following pseudo first-order kinetic equation

$$\frac{dc_{fish}}{dt} = k\left(Tc_{water} - c_{fish}\right)$$
$$k = \begin{cases} k_{up} & \text{when } Tc_{water} \geq c_{fish} \\ k_{ex} & \text{when } Tc_{water} < c_{fish} \end{cases} \qquad (2)$$

where c_{fish} and c_{water} are the ^{137}Cs concentrations in fish (Bq kg^{-1}) and water (Bq m^{-3}), respectively, T is the transfer factor in the equilibrium state (m^3 kg^{-1}), k is the kinetic rate (yr^{-1}). Different kinetic rates were adopted for uptake, k_{up}, and excretion, k_{ex}. c_{water} was obtained by dividing the total ^{137}Cs inventory in river water by the total river water volume. It was assumed that ^{137}Cs uptake by fish did not alter the mass balance in the rest of the system.

2.2 Modelling of generic Fukushima-type forest catchment

In this study we modelled a generic river catchment by using parameters with general applicability for contaminated forest catchments in Fukushima Prefecture. The model included only forest and river land uses, i.e. no urban, paddy or grass areas were included. The ratio of deciduous to coniferous forest by area was assumed to be 4 to 1, in accordance with the average for Fukushima Prefecture measured from a land use map (Japan Aerospace Exploration Agency, 2018). The ratio of river to the forest area was assumed to be 0.01, based on the land use map. In order to calculate the total river water volume, a mean river water depth of 0.4 m was assumed, based on field measurements of rivers such as the Ukedo River in Fukushima Prefecture (Onishi et al., 2014). The annual precipitation was assumed to be 1200 mm yr^{-1} and the ratio of runoff to precipitation was 0.6, based on the average for Fukushima Prefecture (Japan Meteorological Agency, 2018; Unoki, 2010).

Inventories of ^{137}Cs in each compartment were normalized by the total inventory per unit area of the catchment for all calculations. Thus, the results are independent of total ^{137}Cs inventory of the catchment. The model assumes a homogeneous ^{137}Cs distribution over the catchment, i.e. constant ^{137}Cs radioactivity per area. The normalization means the results are also independent of the catchment size.

2.3 Setting of transfer parameters and initial state of model

The main parameters in the model are the transfer rates λ_{ij} between the compartments, the ^{137}Cs transfer factor between fish and river water T, and its associated rate constant k. The initial state of the model was the relative radioactivity of each compartment on March 15, 2011. The following sections describe the way the parameters and the initial state of the model were set. In brief, the transfer rates between forest compartments were obtained by

fitting monitored ^{137}Cs concentrations within four forests in Fukushima Prefecture between 2011-2017. Transfer rates for the forest to river water pathways (direct litter fall, lateral inflow from litter and from soil) were investigated by numerical exploration of the parameter space. Transfer parameters for ^{137}Cs uptake by freshwater fish were assigned based on measured ^{137}Cs concentrations of freshwater fish and river water. The transfer rate from rivers to the downstream compartment was fixed to ensure the overall mass balance of water in the system.

2.3.1 Inverse analysis to estimate fallout interception ratios and transfer rates in forests

The initial ^{137}Cs inventories and transfer rates for forest compartments were estimated by fitting monitoring results from one deciduous forest (konara oak, *Quercus serrata*) in Otama village (OT-Q), and three coniferous forests in Kawauchi village in Fukushima (Ministry of Agriculture, Forestry and Fisheries, 2018b). The Kawauchi coniferous forests were the hinoki cypress (*Chamaecyparis obtusa*) forest, KU1-H, and Japanese cedar (*Cryptomeria japonica*) forests, KU1-S and KU2-S (Imamura et al, 2017). As the goal of the study was to examine the ^{137}Cs transport pathways from forests affecting the ^{137}Cs concentration of freshwater fish, it was considered reasonable to use a backwards fitting approach to obtain the transfer parameters pertinent to internal cycling in the forests. An approach based on direct measurements of the transfer rates was not considered feasible.

The atmospheric fallout at March 15, 2011 was assumed to be the sole input of ^{137}Cs to the system. No inventory was applied to sap wood, heart wood and soil beneath the litter layer in the initial state. The relative interception of the ^{137}Cs fallout by the leaf/needle, branch, bark and litter compartments and the transfer rates between the forest compartments were obtained by minimizing the objective function

$$f = \sum_i w_i \left(\log y_{sim,i} - \log y_{meas,i} \right)^2 . \quad (3)$$

Here $y_{sim,i}$ and $y_{meas,i}$ are respectively the simulated and measured relative ^{137}Cs concentrations in compartment i at time t (i = leaves/needles, branch, bark, sap wood, heart wood, litter layer and soil), and w_i are weights that were chosen manually as to obtain reasonable fitting results.

The objective function was minimized using an iterative process whereby a ±4% alteration was made to one parameter at a time. If the change resulted in f decreasing, the changed parameter was accepted. The iterative process of testing trial changes to the parameters continued until no further changes were acceptable and the process had converged. During this fitting process, the outward flux of ^{137}Cs from forests was neglected.

The initial interception ratios and forest compartment transfer rates estimated by the inverse fitting analysis were cross-checked against an independent set of results from Kato et al. (2017, 2018b). The Kato et al. results are initial interception ratios and radiocesium transfer fluxes from the tree canopy to the forest floor in a mixed-broadleaf forest, mature cedar forest, and a young cedar forest in Kawamata town in Fukushima.

2.3.2 Numerical exploration of transfer parameters from forests to rivers

No appropriate measurements were available for the relative importance of the three transfer pathways in the model from forests to rivers. Thus we investigated the transfer parameters for these pathways numerically by exploring the parameter space with various simulation cases (Table 2). It was assumed that the transfer rates for the three compartments connected to rivers were constants.

In the first three cases (Case 1 to 3) a single transport pathway was assumed: Case 1 – only lateral inflow from soil; Case 2 – only lateral inflow from the litter layer; and Case 3 – only direct litter fall into rivers. The transfer rate for each case was estimated from the bulk

transfer rate from forests to rivers $\lambda_{forest\_to\_river}$ (yr^{-1}) at around two years after the fallout. The bulk transfer rate at this time was obtained as the ratio of the annual ^{137}Cs discharge via the catchment outlet to the total ^{137}Cs inventory in the catchment:

$$\lambda_{forest\_to\_river} = \frac{c_{water} \times R}{I} \quad . \qquad (4)$$
$$R = P \times a \times r$$

Here R is the annual runoff ($m^3\ yr^{-1}$), I is the ^{137}Cs total inventory in the catchment (Bq), P is the annual precipitation P = 1.2 (m yr^{-1}), a is the catchment area (m^2) and r is the ratio of runoff to precipitation r = 0.6 (dimensionless). The bulk transfer rate $\lambda_{forest\_to\_river}$ changes over time but can be estimated at certain time points. Yoshimura et al. (2015) showed that the dissolved ^{137}Cs concentration of river water (Bq m^{-3}) was related to the average catchment inventory (Bq m^{-2}) at around two years after the deposition by

$$c_{water} = 8.6 \times 10^{-5} \times \frac{I}{a} \, . \qquad (5)$$

Thus we can derive

$$\lambda_{forest\_to\_river} = 6.2 \times 10^{-5} \ yr^{-1}. \qquad (6)$$

The ratios of ^{137}Cs inventories in forest compartments for two years after the deposition were roughly 60% in soil, 30% in litter layer and 0-5% in leaves/needles, thus we set the transfer rate from soil to river in Case 1 as 6.2×10^{-5} / 0.645 x 1.5 = 1.4×10^{-4} yr^{-1}, the transfer rate from litter layer to river in Case 2 as 6.2×10^{-5} / 0.345 x 1.5 = 2.7×10^{-4} yr^{-1}, and the

transfer rate from leaves/needles to river in Case 3 as 6.2×10^{-5} / 0.01 x 1.5 = 9.3×10^{-3} yr^{-1}, respectively. Here, the values of 0.645, 0.345 and 0.01 were assumed instead of 0.6, 0.3 and 0-0.05 so that the sum total is 1.0.

The value of 1.5 was added as a tuning parameter. Tuning was necessary because the ^{137}Cs concentrations in river water measured by Yoshimura et al. (2015) were about 1.5 times smaller than the more comprehensive measurements from the Ministry of Environment (2018) used to validate the compartment model. Since this study focuses on the time trend of ^{137}Cs in water and fish rather than the absolute values of ^{137}Cs concentrations, and the value of 1.5 itself is not significant considering the uncertainties in the other parameters, this tuning step is not considered to affect the conclusions of this study.

Cases 4-6 allowed multiple pathways for ^{137}Cs export from forests to river (Table 2). Case 4 permitted transfers from the leaf/needle and litter layer compartments to river water. The transfer parameters used meant each transfer pathway contributed 50% of the total discharge flux from forests to rivers at the two year time point (i.e. on March 15, 2013). No flux from the soil was allowed in Case 4. Cases 5 and 6 allowed contributions from all three pathways. The transfer parameters chosen for Case 5 meant that the soil to river water pathway constituted 10% of the total forest to river flux, and the litter fall and litter layer pathways contributed in equal amounts (i.e. 45% each), at the two year time point. The parameters chosen for Case 6 meant the soil to river pathway contributed 50% of the total flux, and the other pathways 25% each, at the two year point.

2.3.3 Other transfer parameters

The transfer rate from the river compartment to downstream compartment $\lambda_{river\_to\_downstream}$ was

$$\lambda_{river\_to\_downstream} = \frac{R}{V_{river}} \qquad (7)$$
$$V_{river} = a \times 0.01 \times 0.4$$

where V_{river} is the total river water volume (m^3). Thus $\lambda_{river\_to\_downstream}$ yields

$$\lambda_{river\_to\_downstream} = 1.8 \times 10^2 \ \ yr^{-1}.$$

Eq. (7) ensures the mass balance of the system.

The transfer factor from river water (Bq m^{-3}) to freshwater fish (Bq kg^{-1}) was assumed to be 1.6 m^3 kg^{-1} based on measured ^{137}Cs concentrations in freshwater fish and dissolved ^{137}Cs concentrations in river water (data from Ministry of Environment, 2018 and Japan Atomic Energy Agency, 2018). The plots in Fig. 3 include data for two different feeding types of fish, masu salmon *Oncorhynchus masou* (resident form), a carnivorous fish, and Japanese dace *Tribolodon hakonensis*, an omnivorous fish. Although previous studies (e.g. Nasvit et al., 2007; Wada et al., 2016a) noted ^{137}Cs concentrations varied between species, there is no obvious difference between two species in the data in Fig. 3. The kinetic rate for transfer from river water to freshwater fish was set based on results from experiments by Fukushima Prefecture (2018c). The Prefecture released uncontaminated fish (masu salmon in resident form) into rivers, and collected them after two days to two months, to measure their ^{137}Cs concentrations. The results indicated a kinetic rate k_{up} of 2.2 - 4.5 yr^{-1}. In the simulations, 3.4 yr^{-1} was adopted which is the middle of this range. The kinetic rate of ^{137}Cs excretion k_{ex} was assumed to be 2.5 yr^{-1}, which corresponds to a 100 day biological half-life.

3. Results and discussions

3.1 Fitting initial interception ratios and the transfer rates within forests

Figure 4 shows the measurement and compartment model results for the partitioning of ^{137}Cs within forests over a 10 year period from March 15, 2011. The results for each compartment are shown relative to the total forest ^{137}Cs inventory. The compartment model data apply after convergence of the fitting process for the initial inventories of the leaf/needle, branch, bark and litter layer compartments and the transfer rates (λ_{ij}) between the forest compartments.

The weights w_i in the objective function (Eq. (3)) were all set as 1 when fitting the measurements for the coniferous forests in Kawauchi (Fig. 4(b) to (d)). For the deciduous forest in Otama (OT-Q, Fig. 4(a)), it was necessary to increase the weights w_i for the leaf, litter and soil compartments to 5. This change was made so that the simulations produced good matches with the measurements for ^{137}Cs concentrations in compartments linked to rivers (cf. Fig. 4(a) and Fig. S1(a), the latter of which shows results with w_i all equal to 1). Note for the Kawauchi forests, acceptable matches were obtained irrespective of whether these weights were 1 or 5 (cf. Fig. 4(b) to (d) and Fig. S1(b) to (d)). Tables 3 and 4 summarize the initial interception ratios and the transfer rates obtained from the parameter fitting process (as per results in Fig. 4).

Similar trends of the relative ^{137}Cs concentrations in forest compartments could be seen for all forest sites in Fig. 4. The ^{137}Cs concentrations in leaves/needles, branches, bark and the litter layer decreased with time, while those in sap wood, heart wood and soil increased to a plateau. At long times the majority of ^{137}Cs within the forests was located within the soil.

Measurements from deciduous forests were only available for the konara oak site in Otama village (OT-Q). Thus the transfer rates for OT-Q were adopted as the parameter set for deciduous forests in the hereafter compartment model simulations. Although a slight difference between the simulations and measurements could be seen for the ^{137}Cs concentration in branches for site OT-Q (Fig. 4(a)), this compartment was not connected

directly to river water thus this difference did not have a significant effect on the following discussions. The plateauing of the results at ~10 years was indicative of equilibrium being reached for internal cycling of the ^{137}Cs within forests.

The internal transfer parameters for coniferous forests were established separately for the three sites where measurement data were available: the hinoki cypress forest (KU1-H) and the Japanese cedar forest plots (KU1-S and KU2-S) in Kawauchi village. The transfer parameters obtained for KU1-S (Fig. 4(c)) were adopted as the reference for coniferous forests in the following simulations, as measurement data were available for within the first year after the fallout for this site only.

We cross checked the initial interception ratios and the transfer rates estimated by the parameter fitting process with independent measurements from Kato et al. (2017, 2018b). Kato et al. (2017) reported that the canopy interception ratios for a mixed forest, a mature cedar forest, and a young cedar forest of 0.23, 0.69 and 0.70, respectively. As shown in Table 3, the interception ratio for the Otama deciduous forest was 0.32, while the results for the Kawauchi coniferous forests were 0.31-0.55. The results suggest that coniferous forests had higher interception ratios than deciduous forests. This is explained by deciduous trees being leafless at the time of the fallout in March 2011. Considering the heterogeneity of the ^{137}Cs distribution in forest (e.g., Kato et al., 2018a; Takada et al., 2016; Imamura et al., 2018), and the uncertainty in the measurements at the different sites, the interception ratios derived by the inverse analysis were in reasonably good agreement with the measurements.

Most transfer rates such as root uptake and translocation inside trees cannot be measured directly. Therefore we only compared internal ^{137}Cs fluxes within forests against measurements of Kato et al. (2018b) of through fall (TF), litter fall (LF) and stem flow (SF) rates (Fig. 5). The measured ^{137}Cs fluxes (Bq m^{-2} yr^{-1}) were normalized by dividing by the deposition inventories for each site (Bq m^{-2}). The combined TF and LF fluxes correspond to

the transfer from leaves/needles and branches to the litter layer in the compartment model. The SF flux corresponds to the transfer from bark to the litter layer in the compartment model.

The graphs in Fig. 5 show large fluctuations between measurement points. The simulated ^{137}Cs fluxes were generally consistent with the order of magnitude of the measurements, hence justifying the transfer rates obtained by the inverse fitting analysis. The estimated transfer rates from leaves/needles to the litter layer shown in Table 4, specifically 0.52-1.3 yr^{-1} for coniferous forests and 2.59 yr^{-1} for deciduous forests, were comparable with the range of 0.15-1.5 yr^{-1} for coniferous forests and 0.14-4.3 yr^{-1} for deciduous forests estimated by Hashimoto et al. (2013). The transfer rates from the litter layer to soil, 0.20-0.67 yr^{-1} for coniferous forests and 0.59 yr^{-1} for deciduous forests, were in an agreement with the Hashimoto et al. (2013) estimates, which were 0.34-6.8 yr^{-1} and 0.34-5.8 yr^{-1} respectively.

3.2 Transfer processes of ^{137}Cs from forests to freshwater fish

After confirming the applicability of the forest parameters obtained by the inverse fitting process, we proceeded to evaluate feasible transfer parameters for ^{137}Cs export from forests to rivers by numerical exploration of the parameter space. Figure 6 shows the results of Case 1 to 3 simulations and measurements for the relative ^{137}Cs concentrations in river water (dissolved) and in freshwater fish (Ministry of Environment, 2018; Japan Fisheries Research and Education Agency, 2017). The measurement samples were collected from the Abukuma River, which flows from south to north through central Fukushima Prefecture, its tributaries, and several rivers in the coastal area of the Prefecture. Measured concentrations of ^{137}Cs dissolved in river water and in fish were normalized by the mean inventory of the area upstream of the locations where samples were taken (using data from the second airborne survey, Nuclear Regulation Authority, 2018; data downloaded from Japan Atomic Energy

Agency, 2018). In Case 1, the relative concentrations of ^{137}Cs dissolved in river water and within fish increased over time. This is because as ^{137}Cs migrated over time into the forest soil layer, this ^{137}Cs became available in the model for export to rivers. The results of Case 1 were not consistent with the measurements, which show a gradual decrease of the relative ^{137}Cs concentrations over time. Therefore ^{137}Cs export from forests due to litter fall into rivers and lateral inflows from the litter layer could not be ignored.

Cases 2 and 3, where only litter layer or litter fall to river fluxes where allowed respectively, showed decreasing relative ^{137}Cs concentrations over time. The rates of decrease were slightly higher than that for the measurements, however. Collectively these results demonstrate that a single pathway cannot explain the measured trend of ^{137}Cs concentrations in freshwater fish.

The results of the simulation cases allowing multiple export pathways are shown in Fig. 7. Case 4 allowed both the litter fall and litter layer transfers to river water. Comparing with Cases 2 and 3, Case 4 yielded results closer to the monitoring data (cf. Fig. 7(a) and (b) with Fig. 6(a) and (b)) for the initial two year period. However for later years Case 4 underestimated the relative ^{137}Cs concentrations. This result suggests that the soil to river water transfer pathway cannot be ignored. This is reasonable as, in later years, the majority of ^{137}Cs within forests is located within soil. Therefore soil to river transfer is likely to be an important contributor of ^{137}Cs input to river water in later years.

In Cases 5 and 6, the transfer pathways from forest soil to river water were switched on. In Case 5, 10% of the total dissolved ^{137}Cs input to river water at the two year time point was attributable to discharge from the soil layer. The results of Case 5 were closest to the measurements (Fig. 7). Case 6 tended to overestimate the monitored ^{137}Cs concentrations for later years. This suggests the transfer rate from soil to river water was too large in Case 6. The above results suggest that the trend of ^{137}Cs concentrations dissolved in river water and within

fish is attributable the changes in concentration levels in leaves, needles, the litter layer and soil in forests over time.

Figure 8 shows the components of the ^{137}Cs fluxes into river water calculated from Case 5. During the first two years, this model indicated the transfer from leaves and needles (direct litter fall) followed by transfer from the litter layer were the dominant processes. There was a cross-over at two years, upon which transfer from the litter layer became most significant until the 4.5 year time point. After 4.5 years, flux from soil became the most important export pathway to river water. The contributions of the three pathways predicted in this model stabilized at around ten years as the ^{137}Cs cycling within forests reaches equilibrium.

It has been reported that there are two or three different characteristic timescales for the decrease in ^{137}Cs concentrations dissolved in river water and within freshwater fish (e.g., Smith et al., 2002; Nakanishi and Sakuma, 2018). Analyses of fallout from atmospheric nuclear weapons testing and the Chernobyl accident suggest that the radiocesium removal rate from catchments is related to soil properties such as clay mineral and organic content (Smith et al., 2002; Spezzano et al., 1993), however these factors were not analyzed in this study. The assumptions and judgements taken in this study yielded a model where the different characteristic timescales are explained by the redistribution of ^{137}Cs that occurs within forests, rather than differences in soil properties. In the initial stage after fallout, the fast rate of decrease of ^{137}Cs concentrations in river water and fish was attributed to the fast decrease of ^{137}Cs concentrations in forest litter, leaves and needles. On the other hand, the long term slower rate of decrease is controlled by the export of ^{137}Cs from forest soil in our compartment model. Since radiocesium is strongly fixed to mineral particles in soil, the organic component in soil is likely to be the most important component for dissolved ^{137}Cs export from the soil layer if our assumptions hold true.

The rate of export of ^{137}Cs from forest soil will likely depend on the distribution of the

^{137}Cs with depth and existing forms within the soil. These factors cannot be accounted in the current compartment model, unless modifications are made, as the soil to river water export rate was assumed to be constant. Previous measurements in Fukushima Prefecture (e.g., Imamura et al., 2017; Nakanishi et al., 2014) have shown gradual migration of ^{137}Cs deeper into forest soil over time, albeit with a slow rate. Studies of the Chernobyl accident and global nuclear weapons fallout (e.g., International Atomic Energy Agency, 2006; Koarashi et al., 2017) reported ^{137}Cs remained mostly in the upper layers of forest soil. Further studies in Fukushima revealed that organic matter in forest soil played an important role in retaining ^{137}Cs (Koarashi et al., 2019; Koarashi and Atarashi-Andoh, 2019). According to our compartment model, affected species of freshwater fish in catchments with forests will continue to take up ^{137}Cs over the long term. This is due to the continuous input of dissolved ^{137}Cs into river water over time from the forest soil layer in the model. However a firm conclusion cannot be drawn as the effects of ^{137}Cs migration into soil and fixation to clay mineral need to be assessed with a more detailed model.

The measured ^{137}Cs concentrations in two fish species with different feeding habits are within an order of magnitude of each other (Fig. 7(b)). This implies that the source of the ^{137}Cs within these fish is basically the same even if the food chains are different. However, the spread of the ^{137}Cs concentrations measured within fish (Fig. 7(b)) is larger than that for dissolved ^{137}Cs concentrations within river water (Fig. 7(a)). This may be due to the wide variety of sizes of individuals and supporting food chains for different species. Simulation Case 5 reproduced the trend of the measurements from six years of monitoring best (Fig. 7). Continued monitoring is needed for future years to check whether the curve predicted by Case 5 is true, and to understand long-term freshwater fish ^{137}Cs concentrations and uptake mechanisms.

This study focused on mountainous catchments covered by forests as these are typical

catchments in Fukushima Prefecture. However, the modeling concept used here, that considers the link between the overland ^{137}Cs behavior and rivers, is expected to be useful for other types of catchment. This is because the chemical components in rivers are always affected by the overland environment.

4. Conclusions

In this paper a compartment model was developed to assess catchment-scale migration of ^{137}Cs and evaluate three potential transport pathways of ^{137}Cs from forests to fish living in mountain streams. Under the modelling assumptions used, the decreasing trend over time of ^{137}Cs concentrations dissolved in river water and within freshwater fish was explained by the decreasing trend of ^{137}Cs concentrations in leaves/needles and litter layer and the increasing trend in organic soil. The fluxes predicted from the model reached equilibrium at around ten years after the initial fallout due to ^{137}Cs circulation within forests reaching a steady state. Modelled reductions in ^{137}Cs concentrations in river water and freshwater fish over the long term are then controlled by the rate of physical decay of ^{137}Cs.

This paper focused only on species of freshwater fish living in mountain streams with short water residence times. In such an “open” system, ^{137}Cs flux from forests controls the contamination levels of the fish. However, for fish in “closed” systems with long residence times, such as lakes, the internal circulation of ^{137}Cs within the system is more complex, and transfers between water, bed sediment and levels of the food chain should be considered. Further field investigations and modelling improvements are required to understand the sources of ^{137}Cs taken up by freshwater fish in closed systems.

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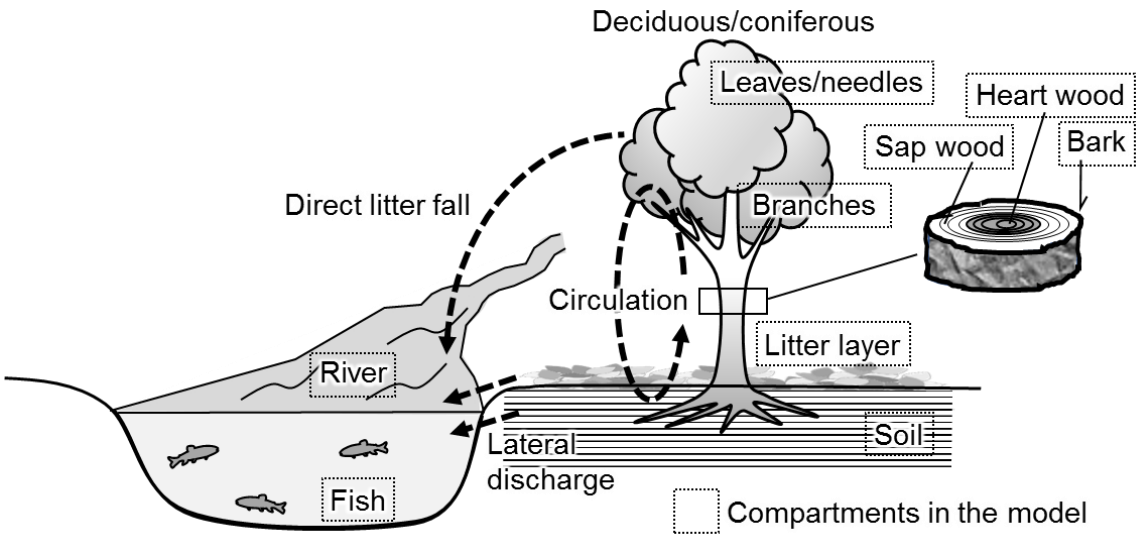

Fig. 1 Schematic of the compartments in the model.

comp., from	to	*j* =1 Leaves/ needles	2 Branches	3 Bark	4 Sap wood	5 Heart wood	6 Litter layer	7 Soil	8-14 *Coniferous forest*	15 River	16 Down-stream	Fish
	Atomo-sphere	intercept.	intercept.	intercept.			intercept.		intercept.	direct fallout		
	Deciduous forest											
i =1	Leaves/ needles	-	translo-cation	weather-ing			litter fall, weathering			direct litter fall		
2	Branches	translo-cation	-	weather-ing	translo-cation	translo-cation	litter fall, weathering					
3	Bark			-	translo-cation		weathering					
4	Sap wood		translo-cation	translo-cation	-	translo-cation						
5	Heart wood				translo-cation	-						
6	Litter layer				root uptake		-	decomp., infiltrate.		lateral inflow		
7	Soil				root uptake			-		lateral inflow		
8-14	*Coniferous forest (compartments same as deciduous forest)*								*same as deciduous forest*	Direct litter fall, lateral inflow		
15	River									-	river flow	uptake

Fig. 2 Interaction matrix of the compartments. Processes listed in each light shaded box represent the transfer processes modelled from compartment *i* to compartment *j*. Blank interactions means there no transfer between those compartments.

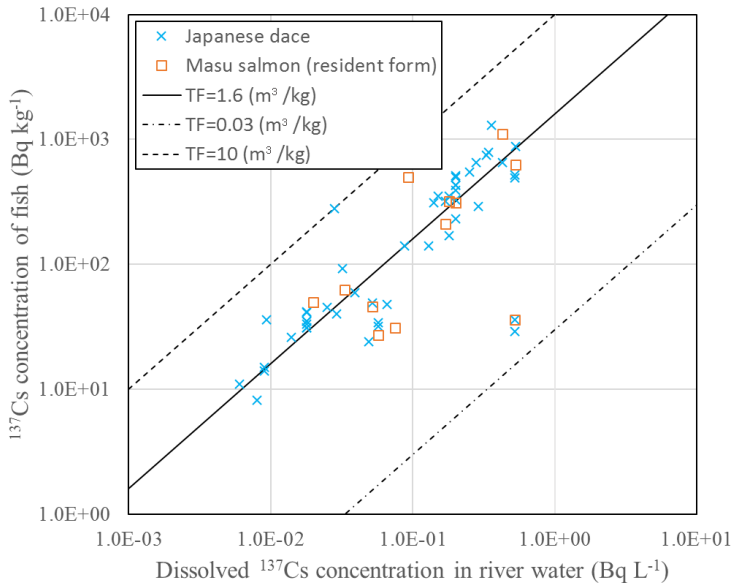

Fig. 3 Relationship between fish and dissovled river water ^{137}Cs concentrations. Measurements by Ministry of Environment (2018).

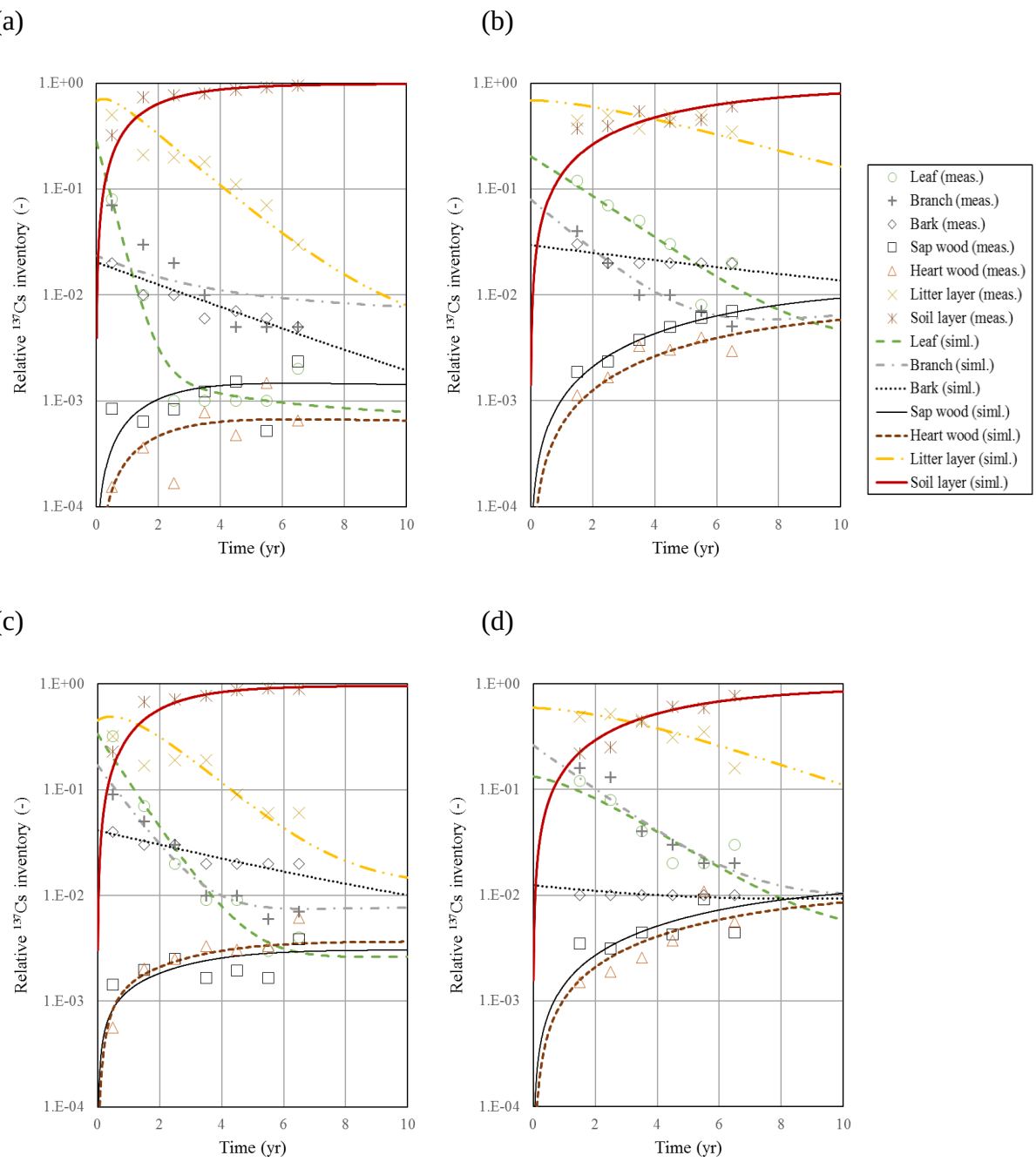

Fig. 4 Relative ^{137}Cs inventory of forest compartments after inverse fitting for transfers parameters and initial state. Graphs show measurements from Ministry of Agriculture, Forestry and Fisheries (2018) (symbols) and compartment model results (lines). (a) Konara oak forest in Otama (OT-Q) – leaf, litter and soil compartment w_i=5. (b) Hinoki forest in Kawauchi (KU1-H), (c) sugi cedar forest in Kawauchi (KU1-S), and (d) second sugi cedar forest in Kawauchi (KU2-S) – all w_i=1.

(a) (b)

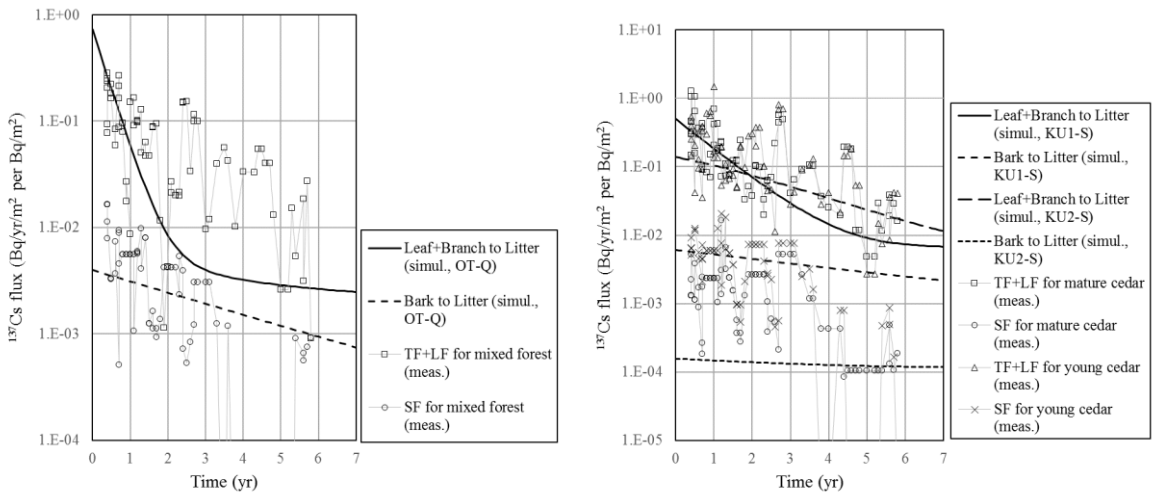

Fig. 5 Comparison of ^{137}Cs fluxes from the canopy to the forest floor between the parametrized simulations and independent measurements by Kato et al. (2018b): (a) simulated konara oak forest at Otama compared with the Kawamata mixed forest, (b) simulated sugi cedar compared with Kawamata cedar forests.

(a) (b)

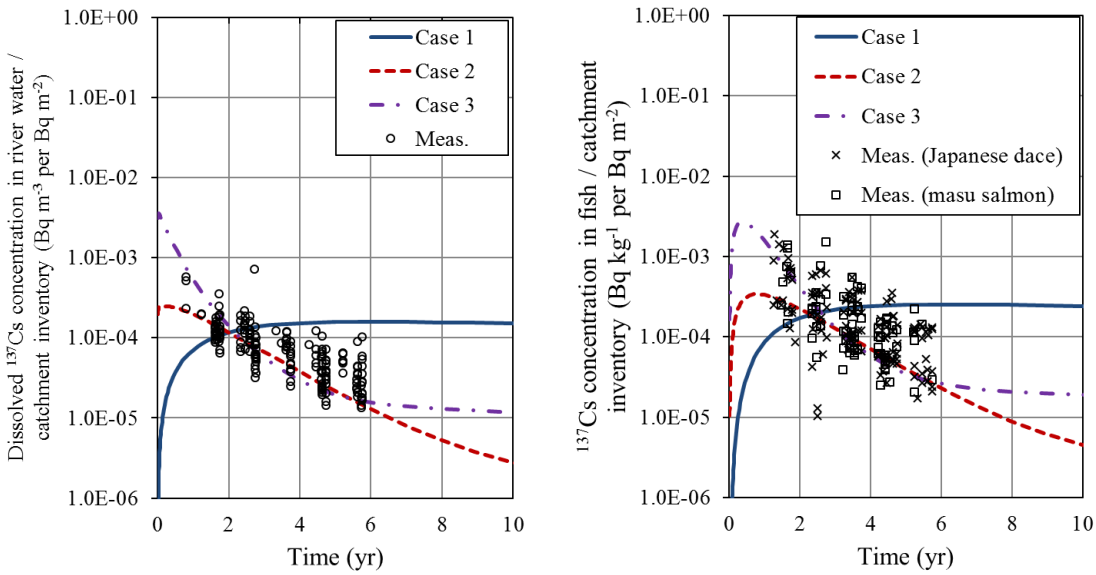

Fig. 6 Simulation results of Case 1 to 3 compared with measurements by the Ministry of Environment (2018) and the Japan Fisheries Research and Education Agency (2017): ^{137}Cs concentrations in (a) river water and (b) freshwater fish.

(a) (b)

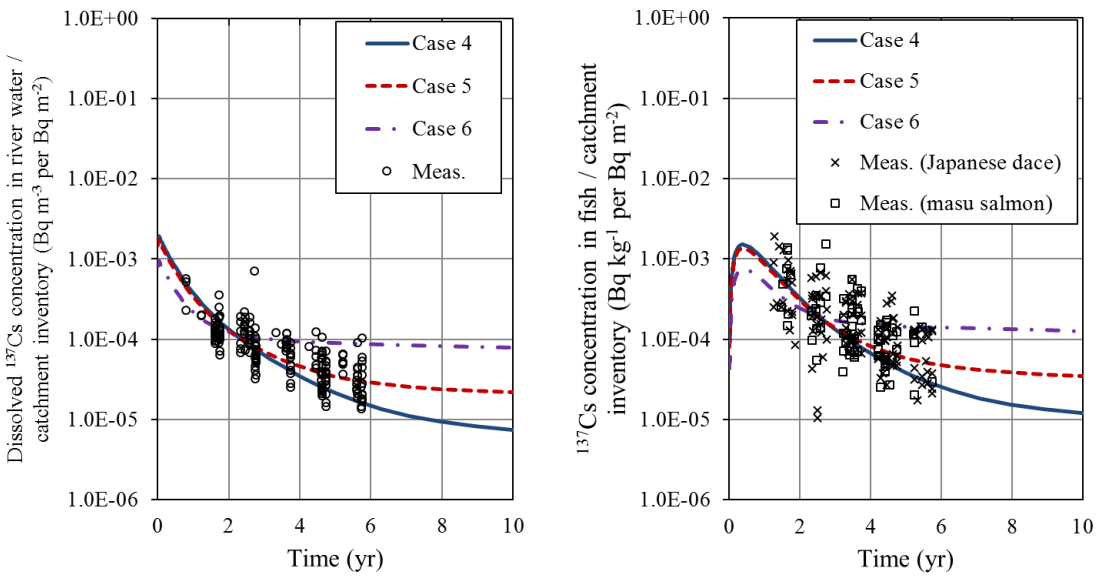

Fig. 7 Simulation results of Case 4 to 6 compared with measurements: ^{137}Cs concentrations in (a) river water and (b) freshwater fish.

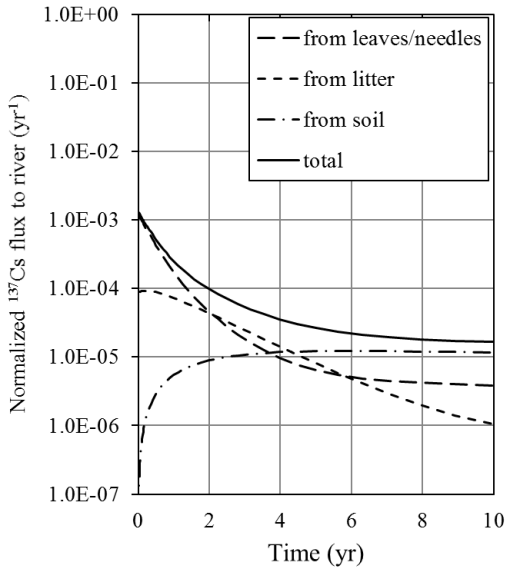

Fig. 8 Normalized relative fluxes of dissolved ^{137}Cs input into river water in Case 5.

Table 1 Concentrations of ^{137}Cs in muscle of major freshwater fish species in Fukushima Prefecture.

Species	Concentration of ^{137}Cs (Bq kg^{-1})	
	Outside the evacuation zone (April 2018 to March 2019) MAFF (2018a)	Inside the evacuation zone (caught in 2016) Fukushima Prefecture (2018b)
Ayu (*Plecoglossus altivelis*)	N.D. - 53.5	Up to 2000
Masu salmon (*Oncorhynchus masou*) (resident form)	N.D. - 126	Up to 20000
White-spotted char (*Salvelinus leucomaenis*)	N.D. - 195	n.a.
Japanese dace (*Tribolodon hakonensis*)	N.D. - 50.4	n.a.
Japanese eel (*Anguilla japonica*)	N.D. - 22.8	n.a.
Common carp (*Cyprinus carpio*)	N.D. - 23.9	n.a.

N.D.: not detected. The detection limit is about 5 to 10 Bq kg^{-1}. n.a.: no applicable measurements.

Table 2 Simulation cases for parameter exploration.

		Transfer rates (yr^{-1}) (percentage of the total discharge flux from forests at two years)		
		From leaves/needles to river	From litter layer to river	From soil to river
Case 1	Single process	0	0	1.4E-4 (100%)
Case 2	Single process	0	2.7E-4 (100%)	0
Case 3	Single process	9.3E-3 (100%)	0	0
Case 4	Multiple processes	4.7E-3 (50%)	1.4E-4 (50%)	0 (0%)
Case 5	Multiple processes	4.2E-3 (45%)	1.2E-4 (45%)	1.4E-5 (10%)
Case 6	Multiple processes	2.3E-3 (25%)	6.8E-4 (25%)	7.0E-5 (50%)

Table 3 Initial fallout interception ratios estimated by inverse fitting analysis.

	Initial interception ratio			
	Deciduous forest	Coniferous forest		
	Konara oak forest in Otama (OT-Q)	Hinoki cypress forest in Kawauchi (KU1-H)	Sugi cedar forest in Kawauchi (KU1-S)	Sugi cedar forest in Kawauchi (KU2-S)
Simulation results				
Leaves/needles	0.28	0.20	0.34	0.13
Branches	0.02	0.08	0.17	0.26
Bark	0.02	0.03	0.04	0.01
Tree total	0.32	0.31	0.55	0.41
Measurement (Kato et al., 2017)	0.23 for the mixed forest	0.69 for the mature cedar forest and 0.70 for the young cedar forest		

Table 4 Transfer rates between forest compartments estimated by inverse fitting analysis.

Transfer process	Transfer rates (yr^{-1})			
	Konara oak forest in Otama (OT-Q)	Hinoki cypress forest in Kawauchi (KU1-H)	Sugi cedar forest in Kawauchi (KU1-S)	Sugi cedar forest in Kawauchi (KU2-S)
Leaves/needles to branches	3.8×10^{-4}	8.0×10^{-3}	2.6×10^{-3}	5.3×10^{-3}
Leaves/needles to bark	5.4×10^{-5}	2.3×10^{-4}	2.1×10^{-3}	1.7×10^{-3}
Leaves/needles to litter layer	2.6	0.52	1.3	0.69
Branches to leaves	0.26	0.29	0.44	0.30
Branches to bark	1.2×10^{-5}	3.3×10^{-4}	1.1×10^{-4}	2.4×10^{-4}
Branches to sap wood	7.2×10^{-4}	4.8×10^{-4}	1.8×10^{-2}	2.5×10^{-3}
Branches to heart wood	1.8×10^{-3}	1.4×10^{-3}	6.7×10^{-3}	2.3×10^{-3}
Branches to litter layer	1.7×10^{-2}	0.29	0.45	0.18
Bark to sap wood	4.9×10^{-2}	4.2×10^{-2}	1.7×10^{-2}	7.8×10^{-2}
Bark to litter layer	0.20	4.2×10^{-2}	0.15	1.3×10^{-2}
Sap wood to branches	1.4	0.44	2.3	0.45
Sap wood to bark	3.6×10^{-2}	2.3×10^{-2}	0.16	8.8×10^{-2}
Sap wood to heart wood	6.0	6.8	4.8	4.2
Heart wood to sap wood	13	11	4.0	5.0
Litter layer to sap wood	1.1×10^{-4}	2.3×10^{-4}	2.7×10^{-5}	7.2×10^{-4}
Litter layer to soil	0.59	0.20	0.67	0.26
Soil to sap wood	1.9×10^{-3}	5.7×10^{-3}	7.4×10^{-3}	6.9×10^{-3}

Supplementary Information to Numerical study of transport pathways of ^{137}Cs from forests to freshwater fish living in mountain streams in Fukushima, Japan

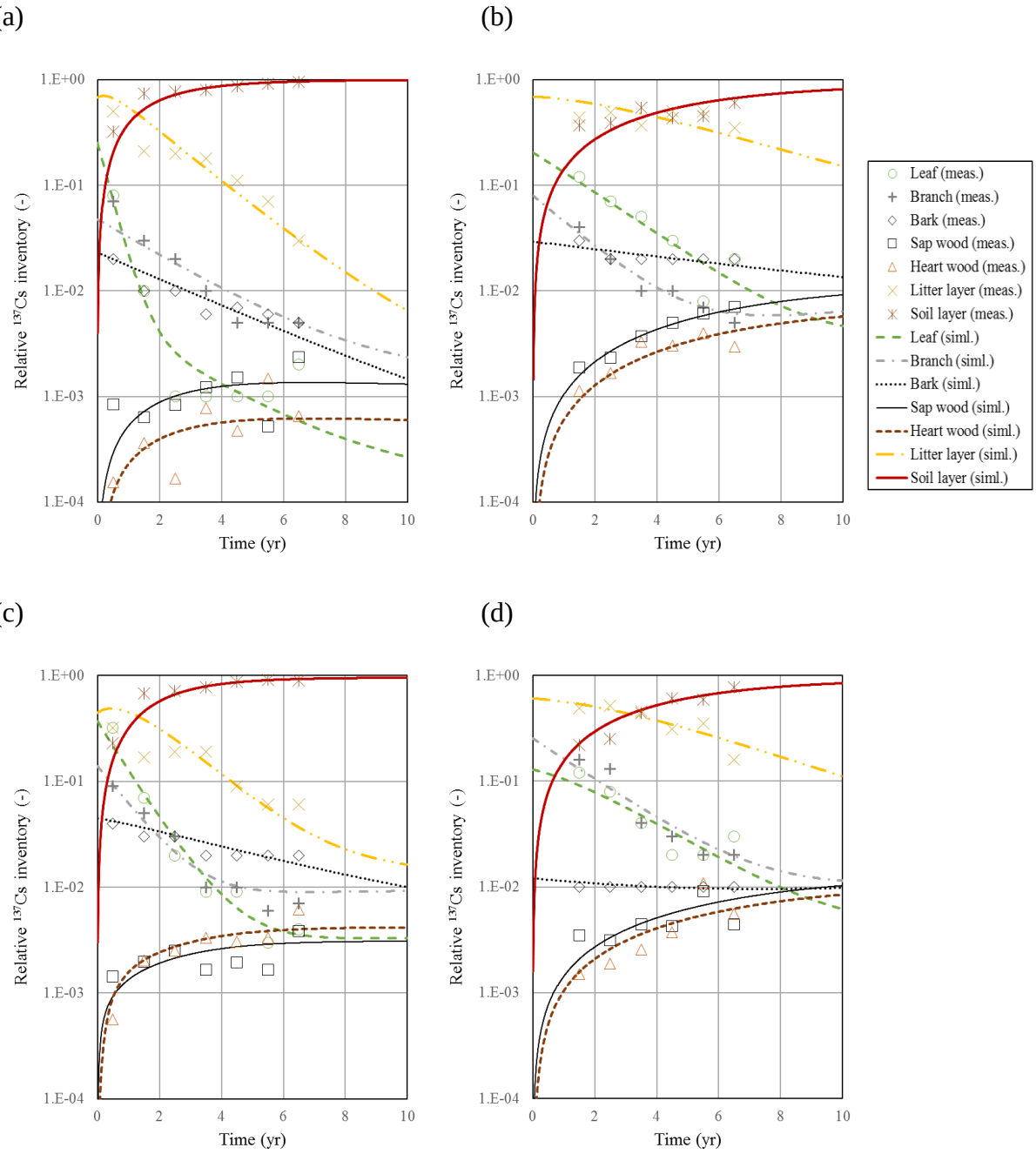

Fig. S1 As per Fig. 4 of main text, but showing compartment results obtained using different weights w_i in the parameter fitting process. (a) Konara oak forest in Otama (OT-Q) – all compartments w_i=1. (b) Hinoki forest in Kawauchi (KU1-H), (c) sugi cedar forest in Kawauchi (KU1-S) and (d) second sugi cedar forest in Kawauchi (KU2-S) – needle, litter and soil compartments w_i=5.